The seasonality of both selenium and vanadium indicates a winter peak (Figure 6-29d). In particular, the vanadium concentration increases by a factor of two for December and January compared to the summer values. Also, the V concentration is higher than over any other region indicating the strongest contribution of fuel oil emissions. The S/Se ratio is strongly seasonal with a winter value of 1,000 and a summer peak of 2,000 to 2,500 consistent with a substantial secondary photochemical contribution of SO_4^{2-} during the summer.

6.4.1.2 Urban Aerosols in the Northeast

In the northeastern U.S. there was a decrease in the annual average PM_{10} concentration between 1988 and 1994 from 28 μg/m^3 to 23 μg/m^3 for all sites and from 31 μg/m^3 to 25 μg/m^3 for trend sites (Figure 6-30b). The reductions were 18% for all sites and 19% for trend sites. The standard deviation among the monitoring stations for any given year is about 30%. The map of the Northeast shows the magnitude of PM_{10} concentrations in proportion of circle radius. The highest AIRS PM_{10} concentrations tend to occur in larger urban centers (Figure 6-30a).

The seasonality of the urban Northeast PM_{10} concentration (Figure 6-30d) is a modest 20%, ranging from 25 to 31 μg/m^3, smaller than the seasonality of the nonurban northwest PM_{10} (Figure 6-29b). There is a summer peak in July, and a rather uniform concentration between September and May showing only a slight winter peak. The $PM_{2.5}$-PM_{10} relationship (Figure 6-30c) shows that on the average 62% of PM_{10} is contributed by fine particles.

In general, the regional scale emissions are not expected to vary significantly from one day to another. However, both meteorological transport (i.e., dilution), as well as aerosol formation and removal processes, are important modulators of daily aerosol concentration. The daily concentration of particulate matter exhibits strong fluctuation from one day to another, mainly due to the role of the meteorological transport variability.

The regionally averaged daily concentration is associated with the regional scale meteorological ventilation. High regionally averaged concentrations indicate poor ventilation (i.e., a combination of low wind speeds and low mixing heights and the absence of fast aerosol removal rates, such as cloud scavenging and precipitation). Low regional concentrations, on the other hand, represent strong horizontal transport, deep mixing heights, or high regional

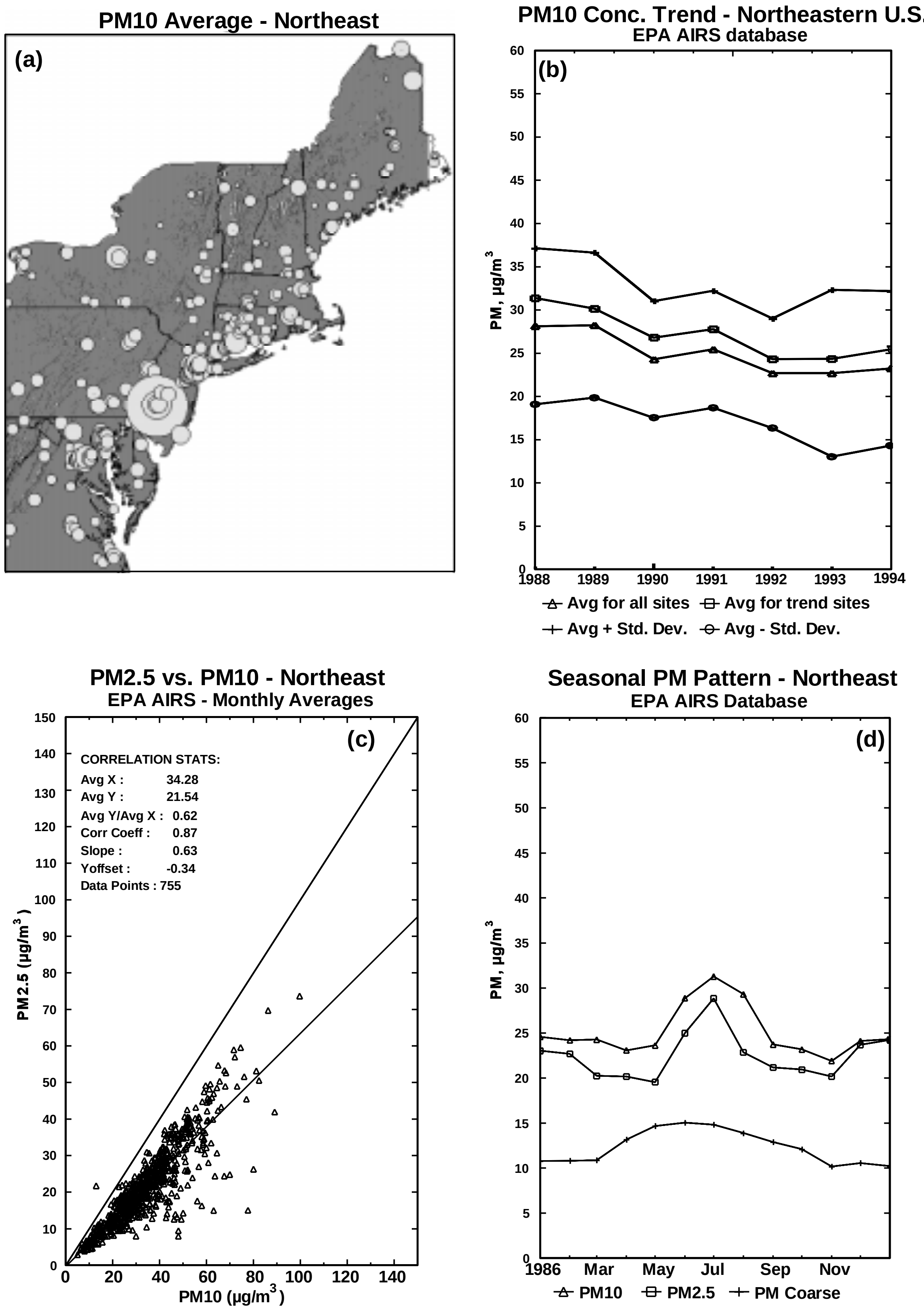

Figure 6-30 AIRS concentration data for the Northeast: (a) monitoring locations; (b) regional PM_{10} concentration trends; (c) PM_{10} and $PM_{2.5}$ relationship; and (d) PM_{10}, $PM_{2.5}$, and PMCoarse seasonal pattern.

removal rates. Advection of high aerosol content air masses from neighboring regions may also be a cause of elevated concentration in a given region.

The daily variation of the regional averaged urban PM_{10} concentration for the Northeast is shown in Figure 6-31. The single day concentration data for every sixth day are connected by a line between the data points, although five in-between days are not monitored. The lowest regionally averaged daily urban PM_{10} is about 10 $\mu g/m^3$, while the highest is about 55 $\mu g/m^3$, with a regional average in the early 1990s of 25 $\mu g/m^3$. The highest concentrations (>40 $\mu g/m^3$) occur primarily in the summer season. The time series also indicate that the high concentration episodes do not persist over consecutive six day periods. This is consistent with the notion that the regional ventilation that is caused by synoptic scale air mass changes, which typically occur every four to seven days over eastern U.S. The daily time series also convey the fact that day to day variation in PM_{10} is higher than the seasonal amplitude. In fact, in Figure 6-31 the concentration seasonality is barely discernible. It can be stated, therefore, that the PM_{10} concentration in the Northeast is highly episodic (i.e., the temporal concentration variation is both substantial and irregular). The excess urban PM_{10} (AIRS-IMPROVE) is shown in Figure 6-32. The excess urban PM_{10} concentration in the Northeast is a relatively small part of the total urban PM_{10} concentration between May and October. The reliability of such estimates of excess regional urban PM_{10} concentrations discussed earlier should be considered (Section 6.3.3).

6.4.2 Regional Aerosol Pattern in the Southeast

The Southeast rectangle stretches from North Carolina to eastern Texas (Figure 6-33). From the point of view of regional ventilation the Southeast terrain is flat, with the exception of the mildly rolling southern Appalachian Mountains. The region is known for increasing population over the past decades, high summertime humidity, and poor regional ventilation due to stagnating high pressure systems.

6.4.2.1 Nonurban Size and Chemical Composition in the Southeast

Only six nonurban stations were available in the Southeast with the absence of monitoring over the southern (Gulf Coast) part of the region, except for Florida. The

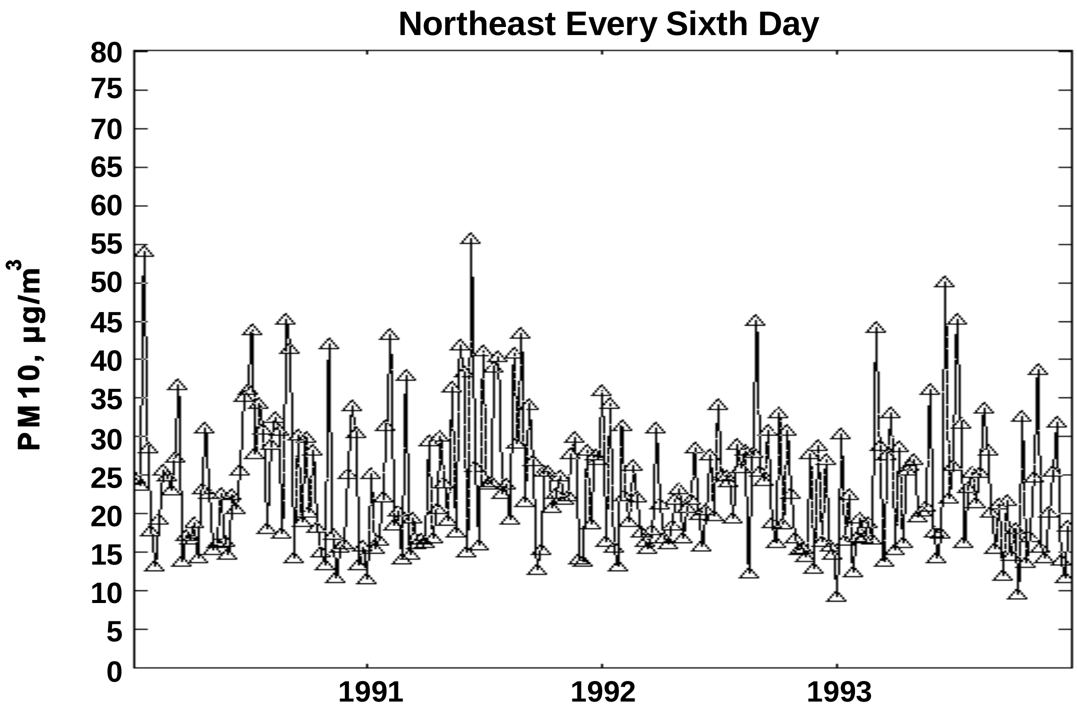

Figure 6-31. Short-term variation of PM_{10} average for the Northeast. Data are reported every sixth day.

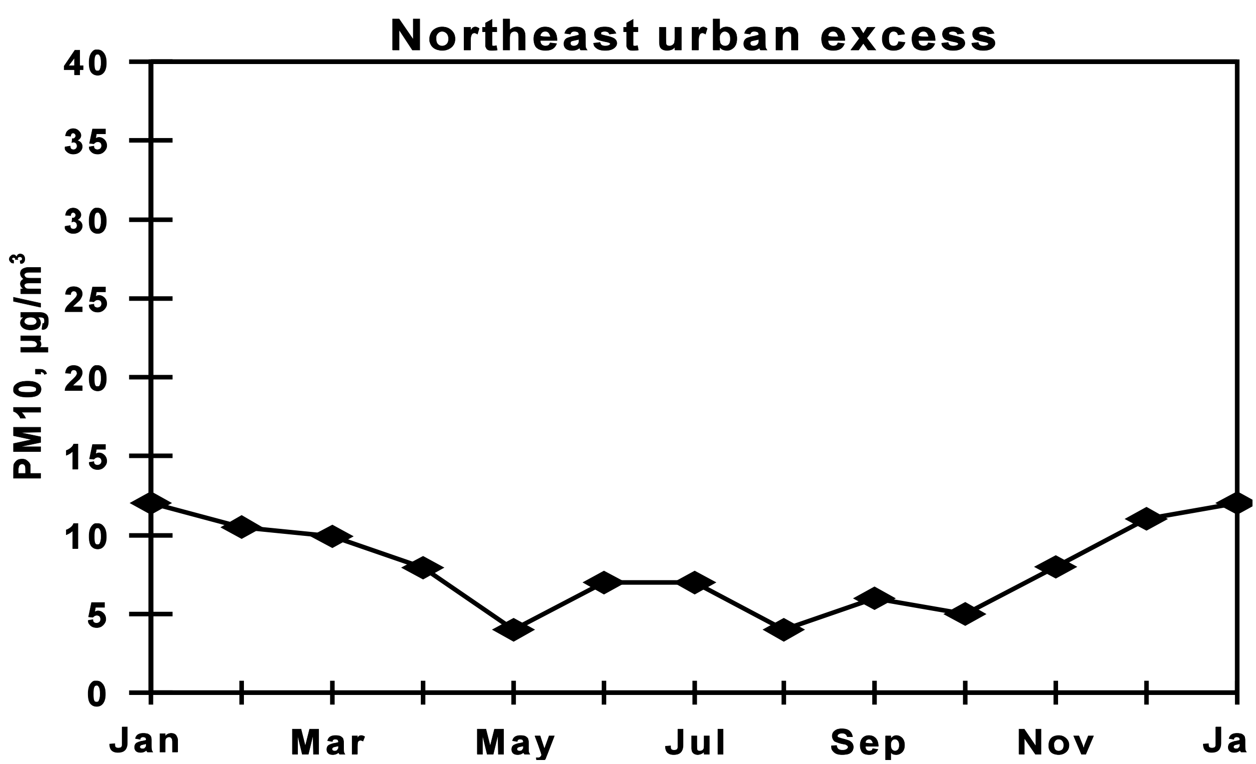

Figure 6-32. Urban excess concentration (AIRS minus IMPROVE) for the Northeast.

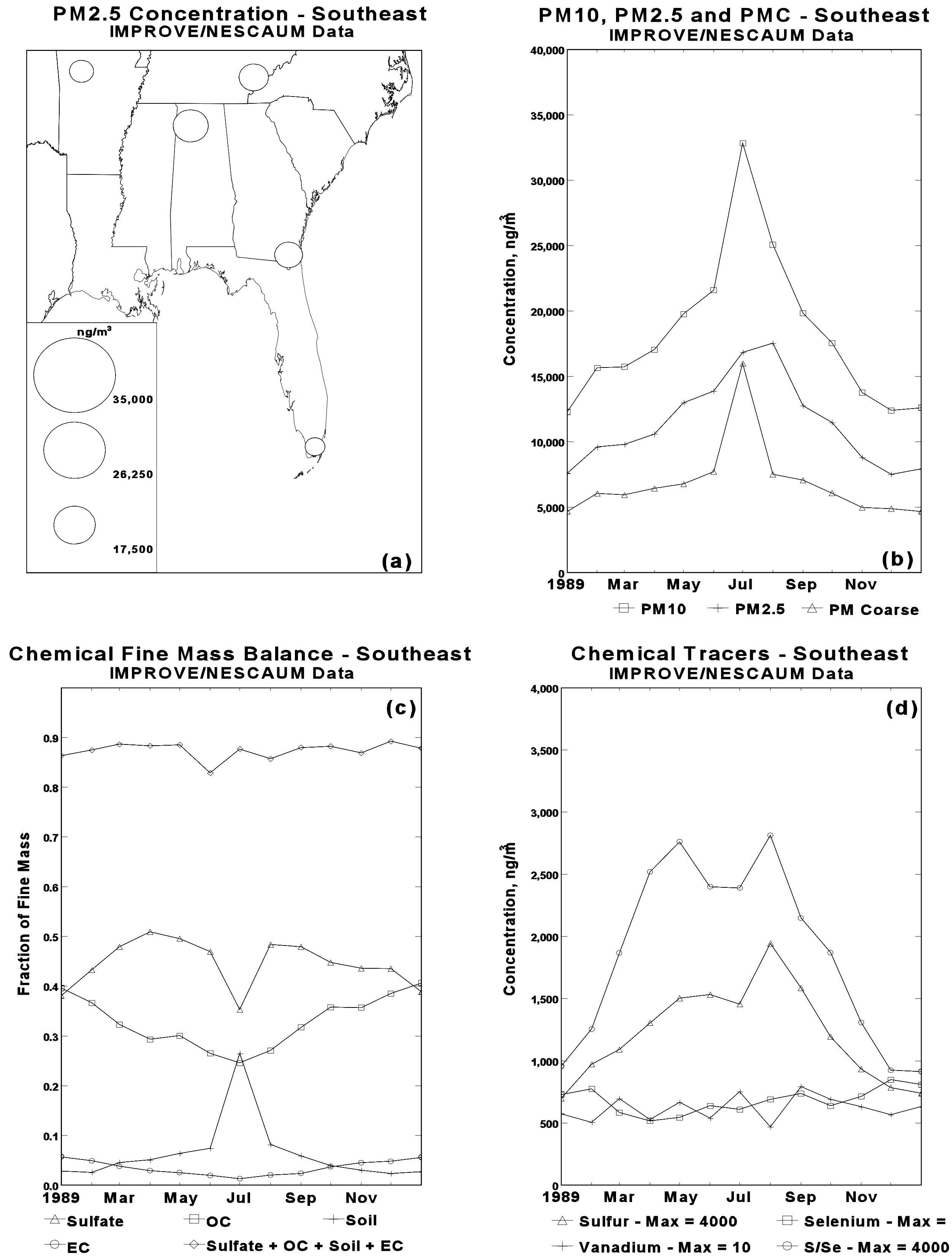

Figure 6-33. IMPROVE/NESCAUM concentration data for the Southeast: (a) monitoring locations; (b) PM_{10}, $PM_{2.5}$, and PMCoarse (PMC); (c) sulfate, soil, organic carbon (OC), and elemental carbon (EC) fractions; and (d) tracers.

nonurban PM_{10} concentration in the Southeast (Figure 6-33b) is roughly comparable to the Northeast, exhibiting about factor of two seasonal concentration amplitude between 12 $\mu g/m^3$ in the winter, and 25 $\mu g/m^3$ in the summer. An anomalous high PM_{10} concentration is shown in July which appears to be contributed by an excess coarse particle concentration of about 10 $\mu g/m^3$. With exception of July, the fine particle mass accounts for about 70% of the nonurban PM_{10}, leaving the coarse mass of 30% or less throughout the year (Figure 6-33b).

The most prominent aerosol species in the Southeast are sulfates contributing 40 to 50% of the fine mass (Figures 6-33c). The anomalously low sulfate fraction (35%) during July coincides with the high (20%) soil contribution during July. For the other months, soil contribution is <5% of the fine mass. The relative role of the organic carbon in the nonurban Southeast is most pronounced during the winter (40%), but declines to 25% during the summer months. The contribution of elemental carbon varies between 2% in the summer to 6% in the winter months.

The trace element concentrations of selenium and vanadium (Figure 6-33d) are constant throughout the year, implying that the combined role of emissions and dilution is seasonally invariant. The concentration of sulfur, on other hand shows a definite summer peak, that is two to three times higher than the winter concentrations. Consequently, the S/Se ratio is strongly seasonal. In fact, the warm season S/Se ratio of 2,500 is higher than over any other region of the country. If Se-bearing coal combustion is the dominant source of sulfur in the Southeast, then the high S/Se ratio implies that the secondary photochemical sulfate production in the summer is several times that in the winter.

6.4.2.2 Urban Aerosols in the Southeast

In the southeastern U.S. there was a decrease in the annual average PM_{10} concentrations between 1988 and 1994 from 33 $\mu g/m^3$ to 27 $\mu g/m^3$ for all sites and from 35 $\mu g/m^3$ to 29 $\mu g/m^3$ for trend sites (Figure 6-34b). The reductions were 18% for all sites and 17% for trend sites. The Southeast PM_{10} concentration trends and the PM_{10} seasonality resemble the industrial Midwest described below. A unique feature of the Southeast is the uniformity of the aerosol concentration among the monitoring stations. In fact the 17% station to station

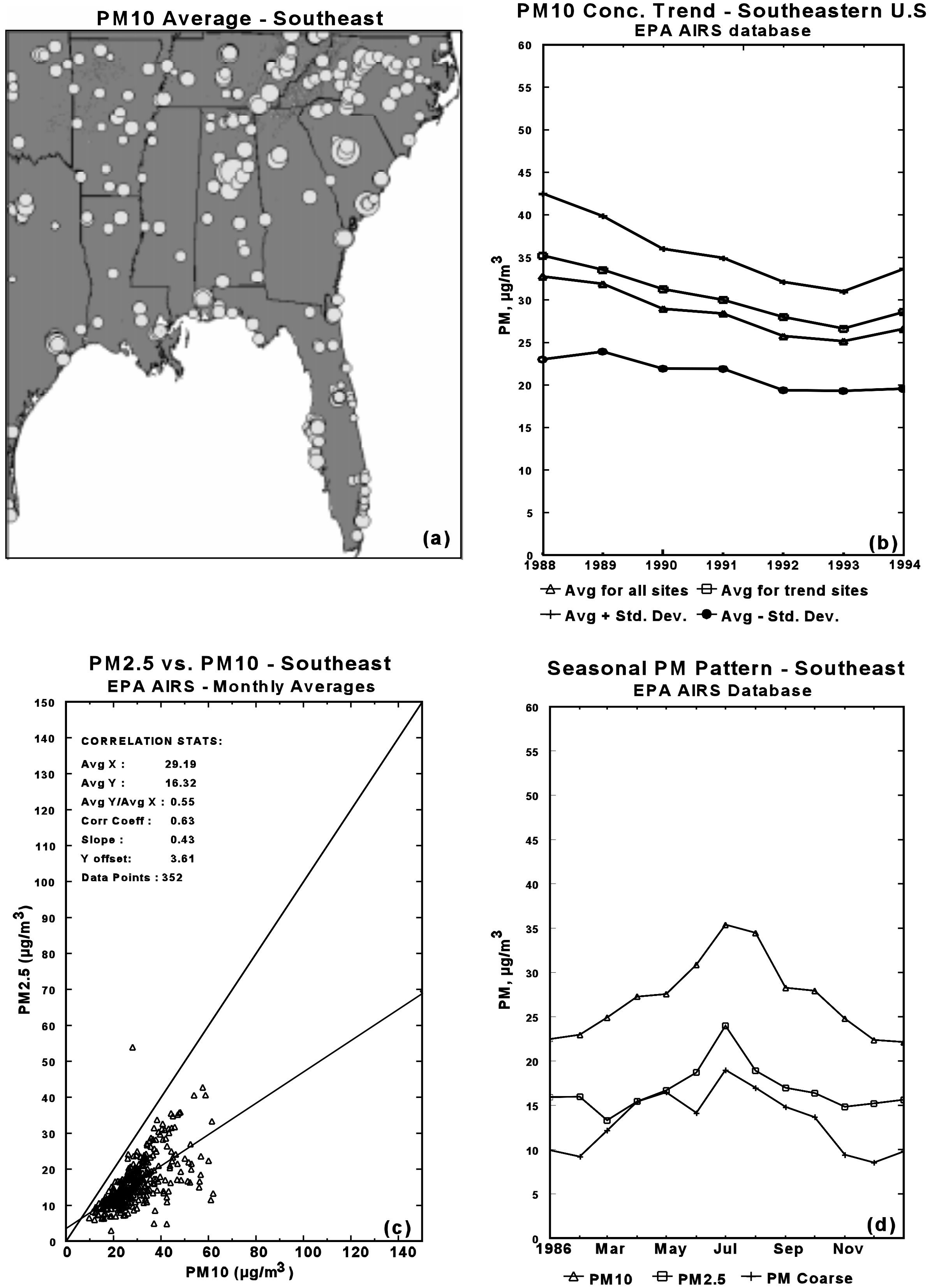

Figure 6-34. AIRS concentration data for the Southeast: (a) monitoring locations; (b) regional PM_{10} concentration trends; (c) PM_{10} and $PM_{2.5}$ relationship; and (d) PM_{10}, $PM_{2.5}$, and PMCoarse seasonal pattern.

standard deviation is by far the lowest among the aerosol regions (Figure 6-34b). This result would appear to be associated with regional meteorological patterns.

The Southeast is also characterized by high seasonal amplitude of 37%, ranging between 22 $\mu g/m^3$ in December through February and 35 $\mu g/m^3$ in July through August (Figure 6-34d). There is no evidence of a winter peak for the southeastern U.S.

The scattergram of $PM_{2.5}$-PM_{10} for the Southeast (Figure 6-34c) shows an average of 58% fine particle contribution, with considerable scatter. It should be noted, however, that size segregated samples were available only briefly and these only for two monitoring sites which may not be representative for the large southeastern region.

The regionally averaged daily PM_{10} concentrations over the Southeast (Figure 6-35) shows a clearly discernible seasonality. The concentrations during the winter months are about factor of two lower than during the summer. Overall, the lowest concentrations are about 12 $\mu g/m^3$, and the highest about 50 $\mu g/m^3$, which is about factor of four. However, seasonality of the temporal signal accounts for about half of the variation. Hence, within a given season the sixth day to sixth day variation is only about 50%**.** It can be concluded that the PM_{10} concentration over the southeastern United States region is quite uniform during shorter time intervals, although it exhibits a substantial seasonality. The southeastern United States also exhibits the highest spatial homogeneity (i.e., the smallest average deviations of average concentrations between the stations). The PM_{10} urban excess (AIRS-IMPROVE) for the southeast region is given in Figure 6-36. The range of monthly urban excess concentrations in the Southeast is within approximately the same range, 5 $\mu g/m^3$ to 10 $\mu g/m^3$, as for the Northeast. The one distinct feature is the sharp decrease in the urban excess in July which corresponds to the sharp peak attributed to the nonurban coarse soil contribution in July for the Southeast (Figure 6-33).

6.4.3 Regional Aerosol Pattern in the Industrial Midwest

This aerosol region stretches between Illinois and western Pennsylvania, including Kentucky on the south (Figure 6-37a). The industrial Midwest is covered by flat terrain west of the Appalachian Mountains. In the winter the region is under the influence of cold Canadian air masses, while during the summer moist air masses transported from the Gulf

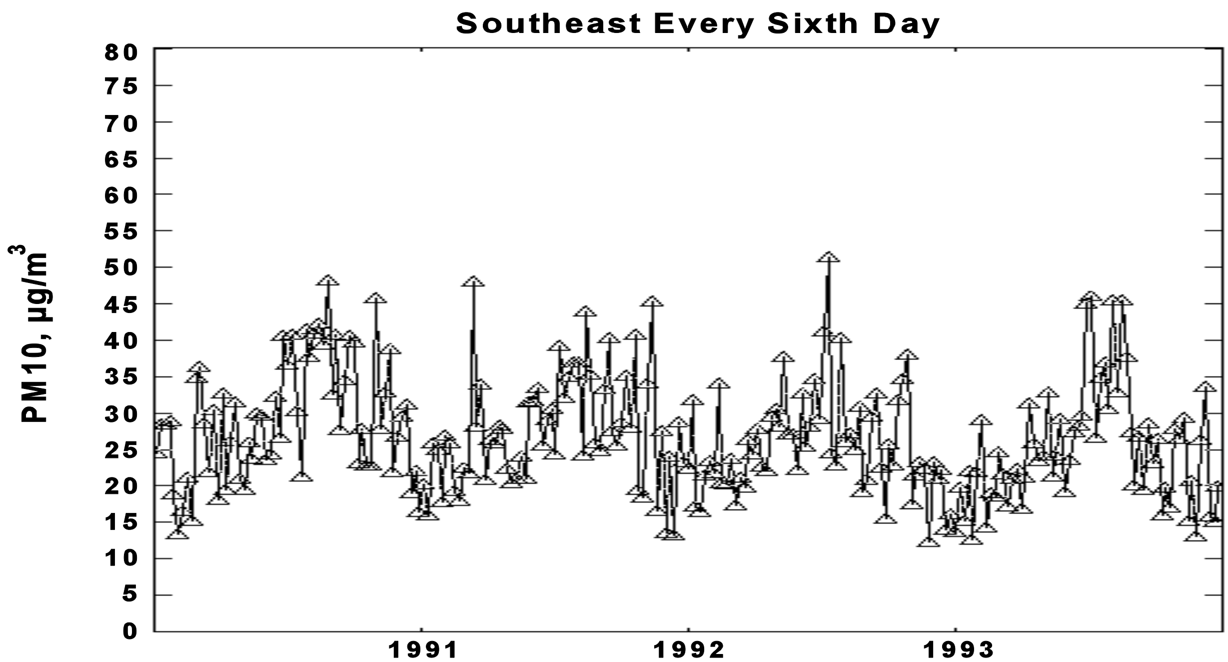

Figure 6-35. Short-term variation of PM_{10} average for the Southeast. Data are reported every sixth day.

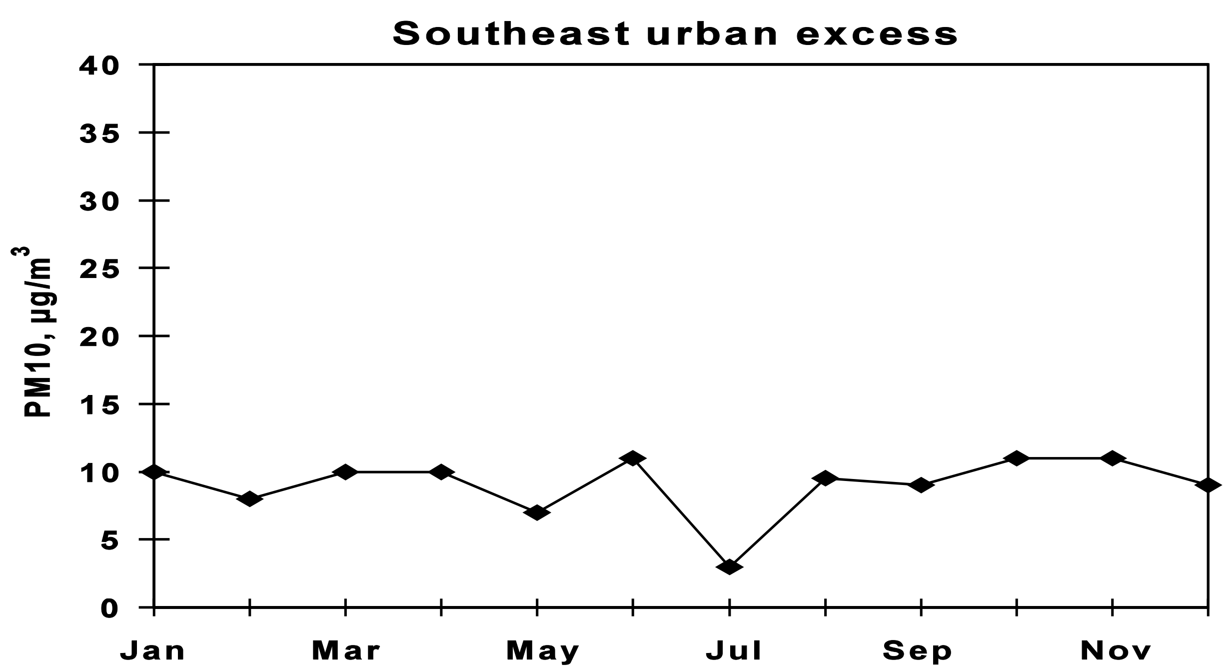

Figure 6-36. Urban excess concentration (AIRS minus IMPROVE) for the Southeast.

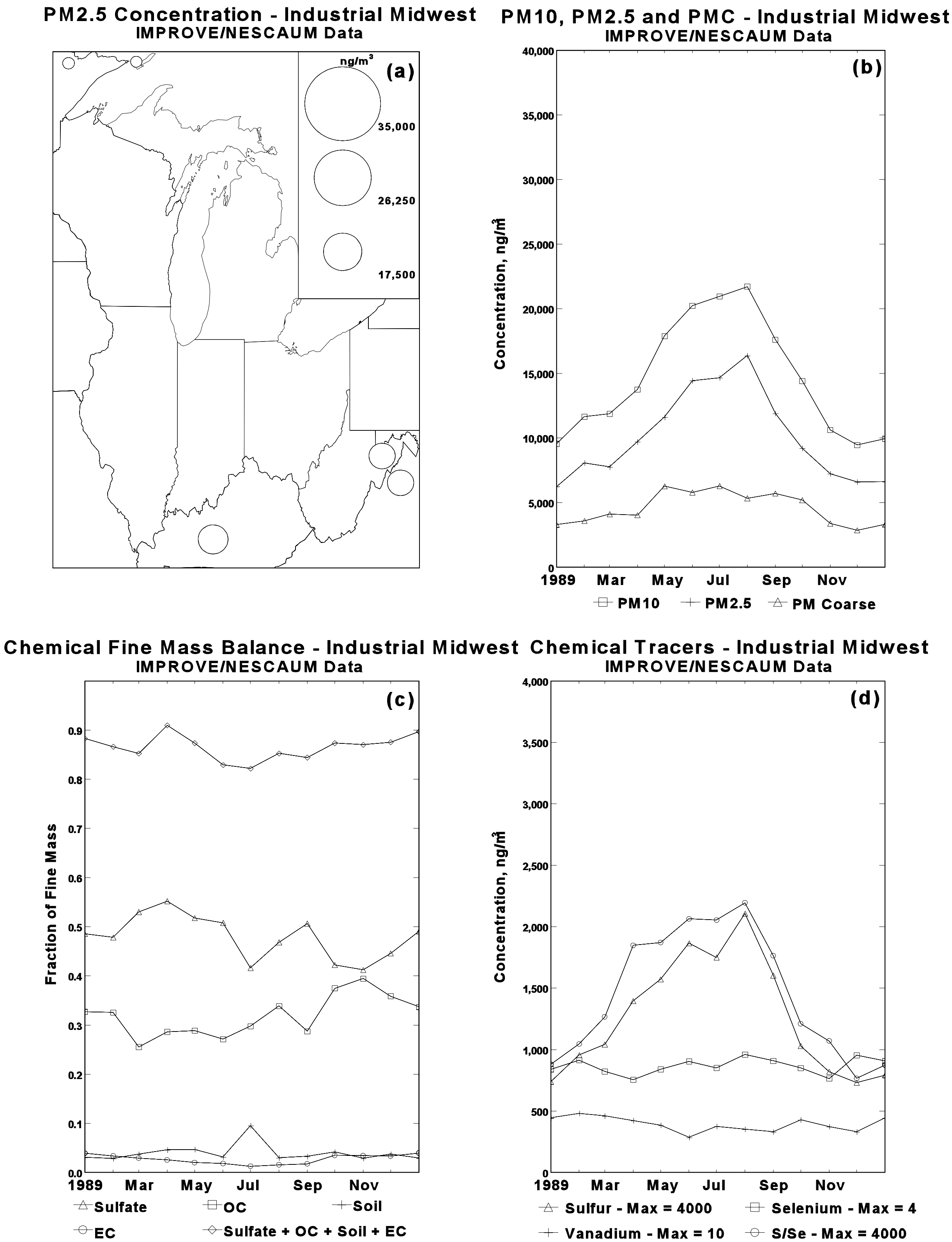

Figure 6-37. IMPROVE/NESCAUM concentration data for the industrial Midwest: (a) monitoring locations; (b) PM_{10}, $PM_{2.5}$, and PMCoarse (PMC); (c) sulfate, soil, organic carbon (OC), and elemental carbon (EC) fractions; and (d) tracers.

Coast prevail. However, the northern most portion of this region in Michigan and Wisconsin is cooler and may be influenced by Canadian air flow at times during the summer. This region includes the Ohio and Mississippi River Valleys that are known for high sulfur emission densities. The region also includes major metropolitan areas.

6.4.3.1 Nonurban Size and Chemical Composition in the Industrial Midwest

The seasonal pattern of the nonurban aerosol in the Industrial Midwest is shown in Figure 6-37b. Only five nonurban monitoring sites are available widely separately geographically between those at the northern most sites and those in the southern portion of the region with no sites over most of the region. Their representativeness is questionable. The PM_{10} concentrations range between 10 and 22 $\mu g/m^3$, comparable to the nonurban levels in other eastern U.S. regions. From 70 to 80% of PM_{10} is contributed by fine particles throughout the year. The coarse particle concentrations are 4 to 5 $\mu g/m^3$, which is lower than over any other region of the U.S. Hence, the contribution of wind blown dust, fly ash, or other man-induced dust entrainment is not a significant factor in the nonurban areas of the Industrial Midwest.

The chemical mass balance (Figure 6-37c) shows that sulfates are 45 to 55% of the fine mass which is higher than the sulfate fractions in other regions. The concentration of vanadium, which is a tracer for oil combustion, is low throughout the year. The concentration of fine particle sulfur Organics exhibit a variable contribution that is high (40%) during the cold season (October through February) and quite low (20%) in July and August. The strong winter peak for the organic fraction differs markedly from the Northeast where the organics are seasonal. Another unusual feature of the chemical mass balance is that the sum of sulfate, organic carbon, soil, and elemental carbon is about 75% during the summer and 95% in the winter. It is not known what is the composition of the missing 25% during the summer time, but the missing fraction could be associated with nitrates, ammonium ion, hydrogen ion, and water.

Chemical tracer data are shown in Figure 6-37d. The chemical tracer for coal combustion, selenium ranges between 1,000 and 1,500 pg/m^3, which is higher than in any other region. There is a sizeable month to month variation in Se concentration (partly due to a small number of data points) and the seasonality is not appreciable. This means that the combined effects of coal combustion source strength and meteorological dilution are seasonally invariant over the industrial Midwest. exhibits random monthly variation but indicates a summer peak. The S/Se ratio is a rather smooth seasonal curve ranging between 1,000 in the winter and 2,000 during the summer months. Hence, the sulfate yield is about twice as high during the summer as during winter months. For comparison both the Northeast and Southeast exhibit somewhat higher seasonality (factor of 2.5) in S/Se ratio. A possible explanation for this change in S/Se ratio is that over the industrial Midwest the average age of the SO_2 emissions traveling downwind may be less than over the Northeast or Southeast.

6.4.3.2 Urban Aerosols in the Industrial Midwest

In the industrial midwester U.S. there was a decrease in the annual average PM_{10} concentrations between 1988 and 1994 from 33 $\mu g/m^3$ to 29 $\mu g/m^3$ for all sites and from 37 $\mu g/m^3$ to 30 $\mu g/m^3$ for trend sites (Figure 6-38b). The reductions were 12% for all sites and 19% for trend sites. There is also a 28% deviation among the stations within the region. As in the Northeast, the higher concentrations occur within the larger urban-industrial areas (Figure 6-38a). The PM_{10} seasonality (Figure 6-38d) is virtually identical (37% amplitude) to the seasonality of the Southeast: the lowest concentrations (25 $\mu g/m^3$) occur between November and February, while the highest values are recorded in June through August (40 $\mu g/m^3$). The trends and the seasonality of the midwestern PM_{10} aerosols are comparable to those of the Southeast.

Fine particles contribute 60% of the PM_{10} concentration on the average (Figure 6-38c), and high PM_{10} can occur when either fine or coarse particles dominate.

Daily concentration over the industrial Midwest (Figure 6-39) varies between 14 and 75 $\mu g/m^3$. The lowest regional concentrations occur during the winter months, while the highest values (in excess of 40 $\mu g/m^3$) occur during the summer. It is evident that seasonality is an important component of the time series, accounting for about half of the variance. The elevated concentrations occur only one sixth day observation at a time, consistent with the low frequency of prolonged episodes. The industrial Midwest also shows substantial spatial variability. The urban excess PM_{10} (AIRS-IMPROVE) for the industrial midwest is given in

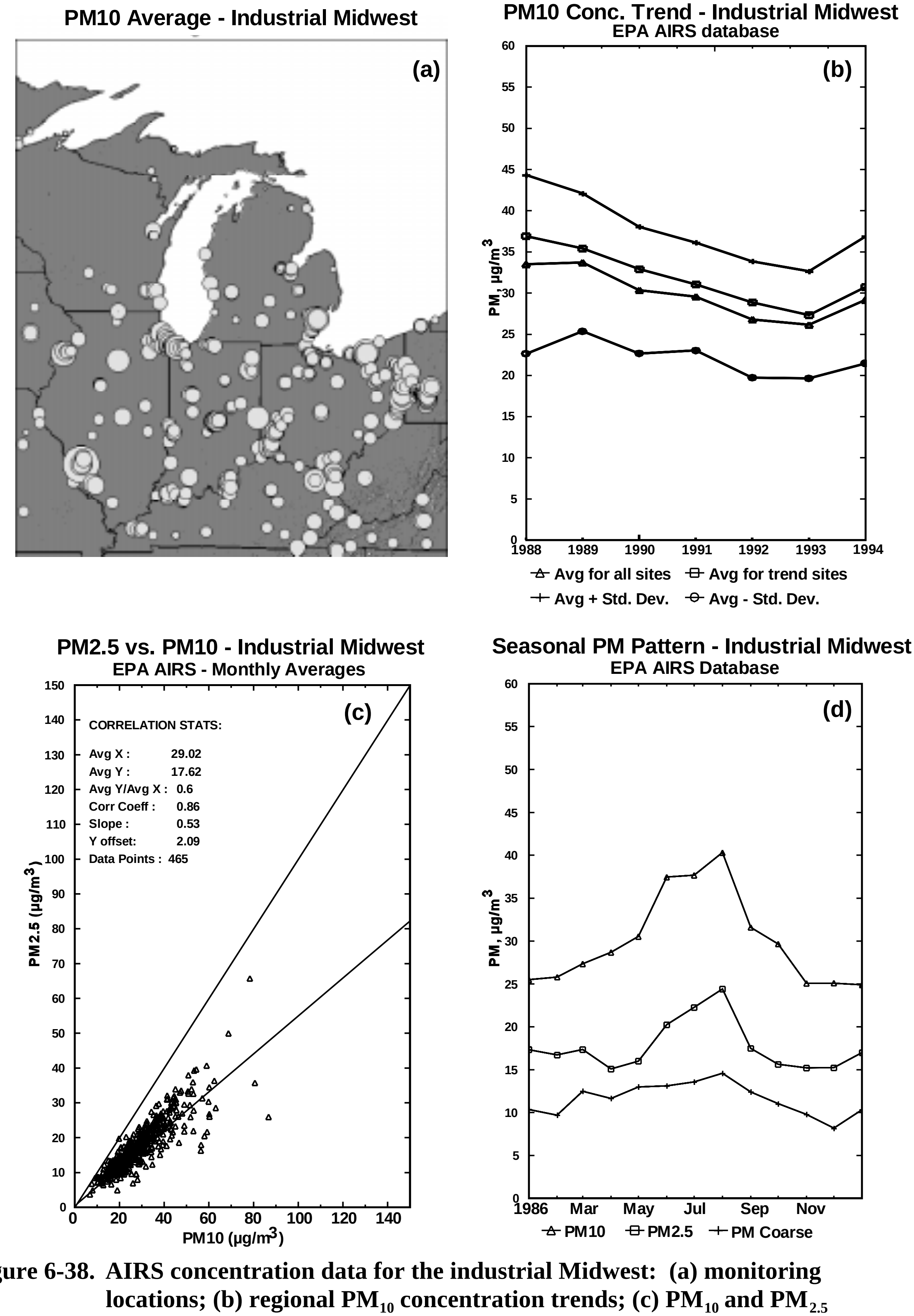

Figure 6-38. AIRS concentration data for the industrial Midwest: (a) monitoring locations; (b) regional PM_{10} concentration trends; (c) PM_{10} and $PM_{2.5}$ relationship; and (d) PM_{10}, $PM_{2.5}$, and PMCoarse seasonal pattern.

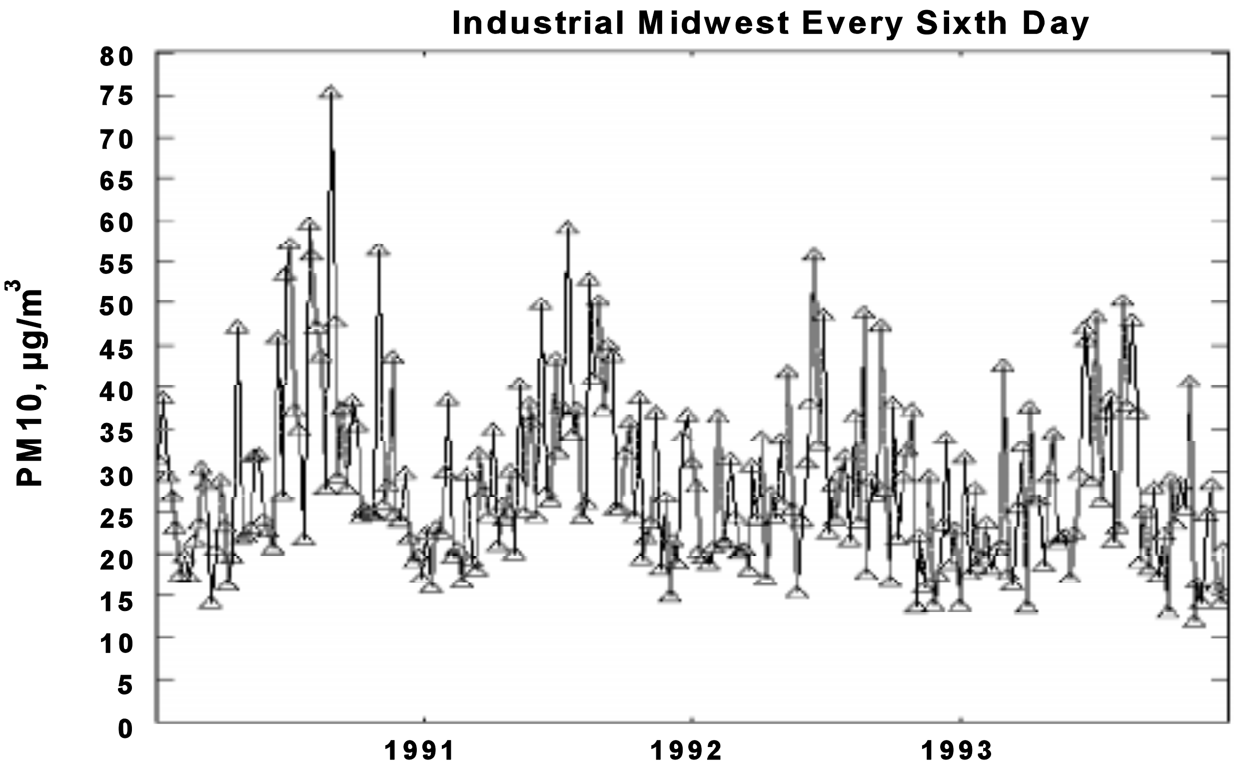

Figure 6-39. Short-term variation of PM_{10} average for the industrial Midwest. Data are reported every sixth day.

Figure 6-40. The pattern for the urban excess PM_{10} differs seasonally from that in the northwest (3-32) or southeast (6-34).

6.4.4 Regional Aerosol Pattern in the Upper Midwest

The upper Midwest covers the agricultural heartland of the country (Figure 6-41). The region is void of any terrain features that would influence the regional ventilation. Industrial emissions and the population density are comparatively low. However, the relatively high PM_{10} concentrations in this region warrant a more detailed examination. In the winter, the region is covered by cold Canadian air masses, while in the summer moist Gulf air alternates and drier Pacific air masses occur.

6.4.4.1 Nonurban Size and Chemical Composition in the Upper Midwest

There is a lack of nonurban monitoring sites in the upper midwest (Figure 6-41a). Compared to the urban sites (Figure 6-42a), these nonurban sites are poorly representative of

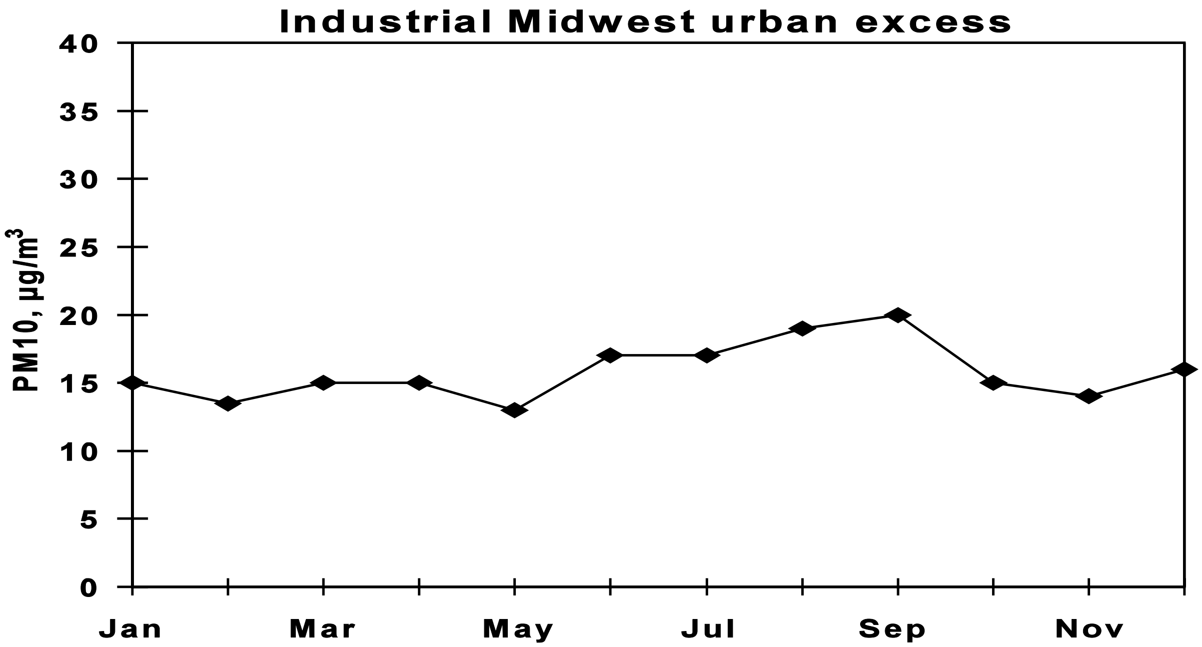

Figure 6-40. Urban excess concentration (AIRS minus IMPROVE) for the industrial Midwest.

the region. Based on these few sites in the upper Midwest, the PM_{10} concentration is about 8 $\mu g/m^3$ during the November through April winter season, and increases to 15 $\mu g/m^3$ during the summer. Fine and coarse particles have a comparable contribution to the PM_{10} mass (Figure 6-41b).

The chemical mass balance (Figure 6-41c) indicates that during the March through May spring season sulfates dominate, but during July through October season organics prevail. This is a rather unusual pattern not observed over any other region. The contribution of fine particle soil exceeds 10% in the spring as well as in the fall season.

Chemical tracers are shown in Figure 6-41d. Selenium concentration is low throughout the year (400 to 600 pg/m^3), with the highest concentrations observed during the summer. This suggests that either the Se sources from coal-fired power plants or the Se transport into the Upper Midwest from other regions is stronger in the summer. The concentration of the fine particle sulfur is <500 ng/m^3 throughout the year, but somewhat higher during March and April. The spring peak of fine particle sulfur has not been observed in any other region. It is also worth noting that S/Se ratio is the highest during the spring and lowest in July

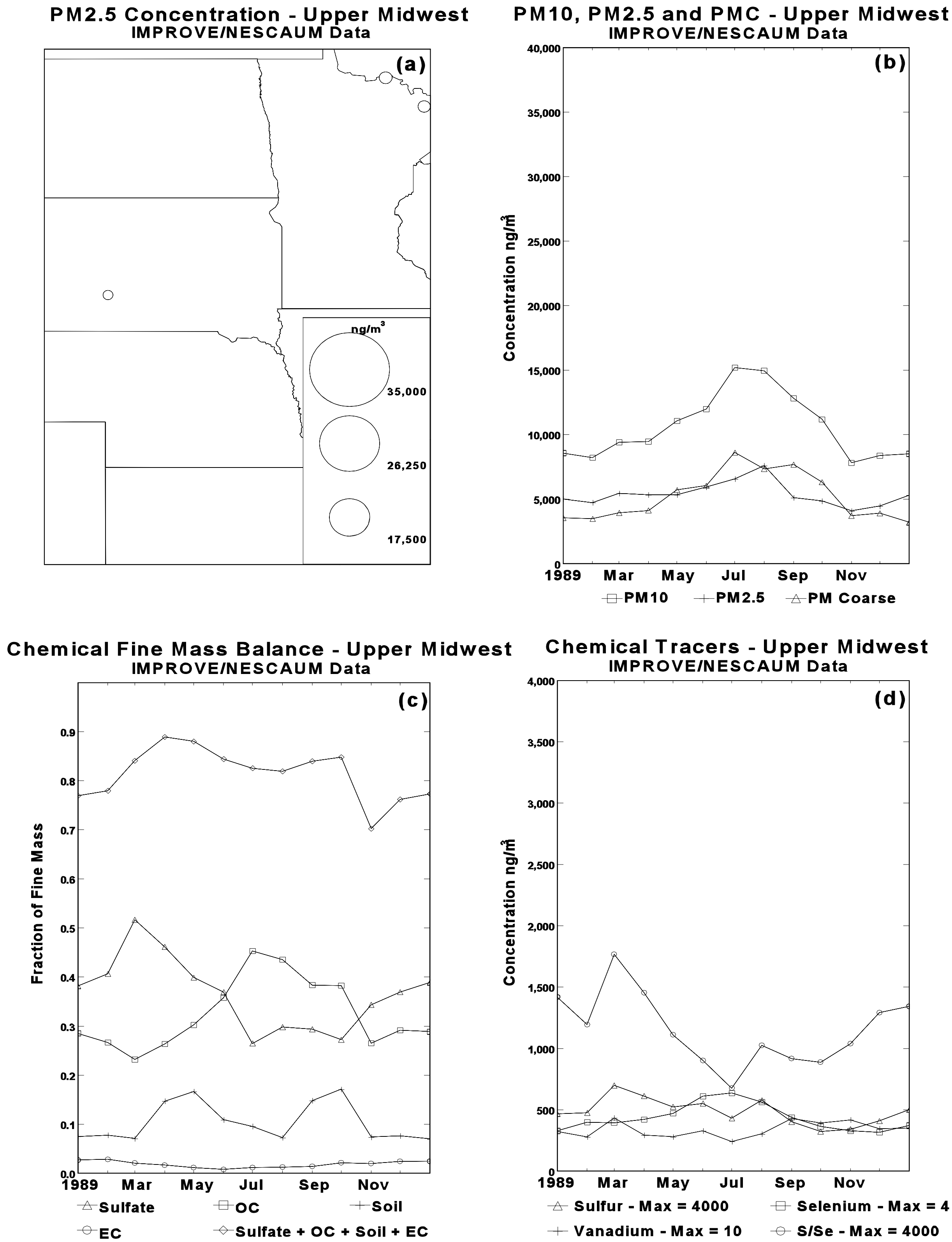

Figure 6-41. IMPROVE/NESCAUM concentration data for the upper Midwest: (a) monitoring locations; (b) PM_{10}, $PM_{2.5}$, and PMCoarse (PMC); (c) sulfate, soil, organic carbon (OC), and elemental carbon (EC) fractions; and (d) tracers.

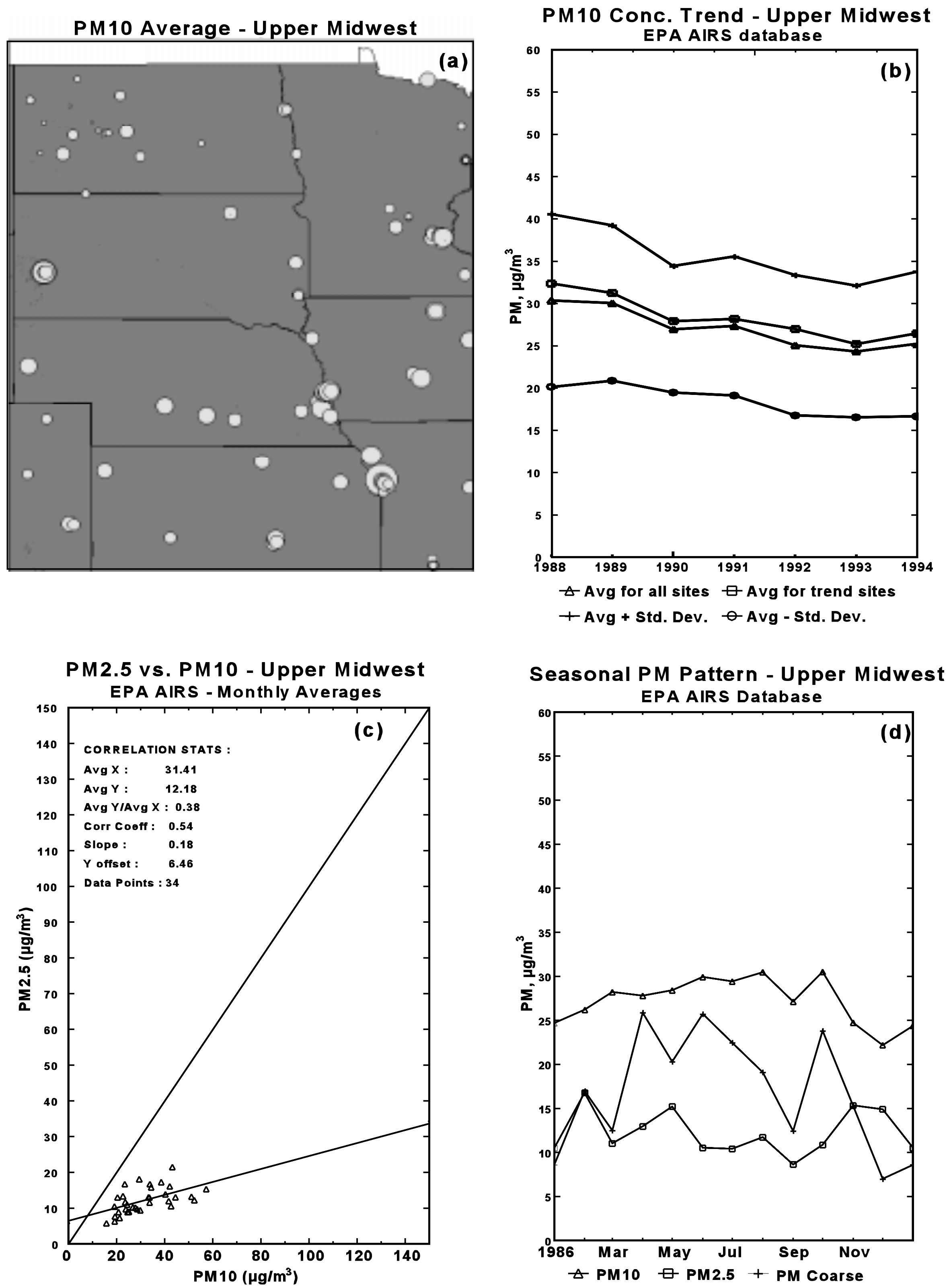

Figure 6-42. Aerometric Information Retrieval System (AIRS) concentration data for the upper Midwest: monitoring locations; regional PM_{10} monitoring trends; PM_{10} and $PM_{2.5}$ relationship; and PM_{10}, $PM_{2.5}$, and PMCoarse seasonal trends.